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Field Induced Superconductivity in a Magnetic Organic Conductor

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Physicists concluded long ago, and after several decades accumulating physical evidence, that superconductivity could not coexist with a magnetic ground state. It is thus easy to understand why the recent discovery of a series of ferromagnetic superconductors, like UGe_2,[1] $ZrZn_2$,[2] and the borocarbides superconductors,[3] has generated such excitement in the scientific community. Here, according to the conventional understanding of superconductivity, the magnetic moments are expected to play a role quite similar to that of an external magnetic field, which destroys superconductivity either by inducing strong diamagnetic currents or by breaking the spin-singlet state of the Cooper pair (the so-called Pauli pair breaking mechanism).

Given our previous understanding of magnetism and superconductivity, it was remarkable that S. Uji and co-authors[4] recently reported clear physical evidence for a magnetic field-induced superconducting state (FISC) in the λ-$(BETS)_2FeCl_4$ layered organic conductor. This observation is particularly unusual since this compound, at zero field and below $T_N = 8.5$ K, is an *insulating* antiferromagnet. According to Uji *et al.*, at low temperatures the FISC state appears only for fields above 18 T and applied along the conducting planes.[4] Motivated by these preliminary results, we recently mapped out the temperature-magnetic field phase diagram of the FISC state[5] by using AC electrical transport techniques in conjunction with the Hybrid magnet of the NHMFL in Tallahassee.

Our magnetic field dependent resistance of a λ-$(BETS)_2FeCl_4$ single crystal, under atmospheric pressure, is shown in Fig. 1(a) for different temperatures. Here, the magnetic field B is applied along the in-plane c-axis. The main characteristic of the data is that between 18 and 41 T, the resistance of the material drops with decreasing temperature, reaching zero within experimental uncertainties below 2 K in a field range centered near 33 T. In the FISC state and at higher fields, the resistivity drops typically by 2 to 4 orders of magnitude, putting it at or below the conductivity of copper, and beyond our ability to measure by standard AC lock-in methods. One of the main results of this study is the observation of reentrance toward the metallic state at a temperature-dependent critical field. Since the presence of Fe^{3+} magnetic moments, coexisting with the FISC state, would suggest a triplet superconducting state that, by definition, is not affected by the Pauli pair breaking mechanism. The fact that the FISC state is re-entrant to a metallic state above 41 T, excludes triplet pairing. Fig. 1(b) shows the resistance of a second single crystal as a function of B (up to 45 T) and for several temperatures. This sample is immersed in a liquid medium that induces a small amount of hydrostatic pressure when it solidifies upon cooling. No pressure was applied at room temperature. Notice that the FISC state is now observable in a much broader field range, indicating that this compound, and hence the FISC state, is remarkably sensitive to pressure.

From the isothermal field scans of Fig. 1, we extracted the temperature dependence of the resistance at fixed values of field (not shown). From this set of data and from Fig. 1, we built the phase diagram shown in Fig. 2. Solid symbols describe the diagram of the FISC state at ambient pressure, while open symbols describe its diagram under $p = \varepsilon$ kbar (>> 1 bar). In both cases, the transition temperature towards the FISC state increases with B reaching a maximum value $T_C = 4.2$ K at $B^* \cong 33$ T for $p = 1$ bar and $B^* \cong 31.5$ T for $p = \varepsilon$ kbar. T_C decreases again for fields above B^* which makes the diagram symmetric around B^*. The solid continuous line is a fit of our data to the so-called Jaccarino-Peter effect.[6] The basic idea is that localized magnetic moments of Fe^{+3} d electrons aligned along the external magnetic interact antiferromagnetically via exchange coupling, with the itinerant π electrons responsible for electrical conduction in this system. The total field $\mu_B H_{eff}$ "felt" by the π electrons in an external field B_0 is thus given by:

$$\mu_B H_{eff} = \mu_B B_0 + J\langle S\rangle \qquad (1)$$

We can clearly see from Eq. (1) that, eventually, a strong enough external magnetic field B_0 can *compensate* the internal exchange field $J\langle S\rangle$ for $J < 0$, allowing the condensation of quasiparticles into a superconducting state if they were subjected to attractive interactions. For more details see References 5 through 7.

Consequently, in the present case, and in contrast to what is observed in conventional superconductors, magnetic moments are the essential ingredient for the stabilization of superconductivity at very high magnetic fields.

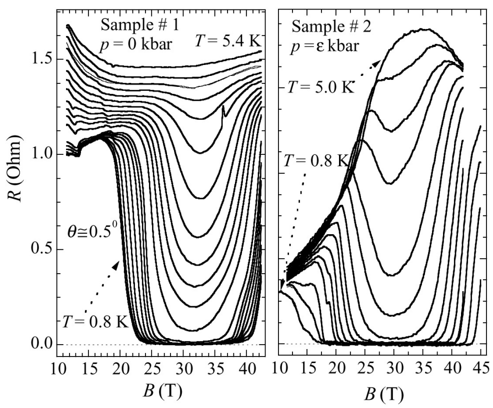

Figure 1. (a) Resistance R as a function of magnetic field B, applied along the in-plane c-axis (± 0.3 degrees) of a λ-$(BETS)_2FeCl_4$ single crystal (sample #1), at ambient pressure and for temperature intervals of approximately 0.25 K, between 5.4 and 0.8 K. The superconducting state develops progressively with decreasing temperature, but is suppressed for fields sufficiently away from (above or below) 33 T. (We note that since the Hybrid magnet is composed of a superconducting outsert coil in combination with a Bitter type resistive insert coil, the field generated by the outsert is kept constant at approximately 11.5 T, while the field of the insert coil was ramped between 0 and 31.5 T). The FISC transition has a maximum transition temperature $T_c \approx 4.2$ K near 33 T. (b) As in (a) R as a function of B, applied along the in-plane c-axis for sample #2. In the present case, the sample is immersed in a fluid medium that induces a very small amount of hydrostatic pressure $p = \varepsilon$ kbar (>> 1 bar) upon cooling. The effect of p is, on one hand, to considerably decrease the resistivity of this compound and, on the other, to widen the range in magnetic fields where the FISC state is observable.

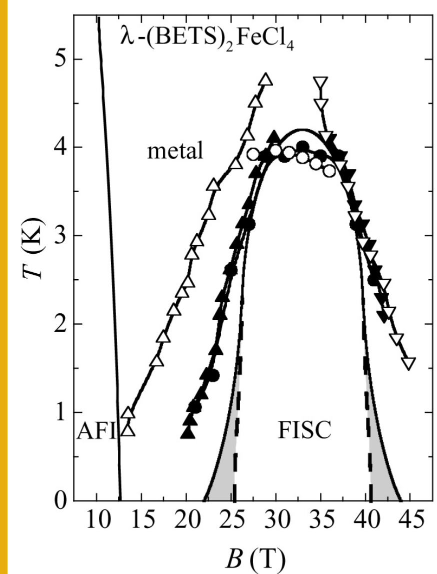

Figure 2. Temperature-magnetic field phase diagram showing the AFI, metallic, and FISC states for a λ-$(BETS)_2FeCl_4$ single crystal vs. in-plane magnetic field at ambient pressure (solid lines and symbols) . Solid triangles indicate the middle point of the resistive transition as a function of B (from Fig. 1(a)), while solid circles indicate the middle point of the resistive transition as a function of T. Similarly, open triangles and open circles describe the T - B diagram of the FISC state under *a very small amount of* hydrostatic pressure (the AFI is displaced to fields B < 8 T). The solid line is a theoretical fit (see text) to a second order phase transition toward the FISC phase while the dashed line indicates a first order transition from the inhomogeneous so-called LOFF state (after Larkin, Ovchinnikov, Ferrell and Fulde)[7] (shaded area) into the bulk superconducting state.

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1. Saxena, S.S., *et al., Nature,* **406,** 587 (2000).
2. Pfleiderer, C., Uhlarz, M., Hayden, S.M., Vollmer, R., Lohneysen, H.V., Bernhoeft, N.R., and Lonzarich, G.G., *Nature,* **412,** 58 (2001).
3. Yaron, U., Gammel, P.L., Ramirez, A.P., Huse, D.A., Bishop, D.J., Goldman, A.I., Stassis, C., Canfield, P.C., Mortensen, K., and Eskildsen, M.R., *Nature,* **382,** 236 (1996). For a review see, Canfield, P.C., Gammel, P.L., and Bishop, D.J., *Physics Today*, 40-46 (1998).
4. Uji,, S., Shinagawa, H., Terakura, C., Terashima, T., Yakabe, T., *et al.*, *Nature*, **410**, 908 (2001).
5. Balicas, L., Brooks, J.S., Storr, K., Uji, S., Tokumoto, M., Tanaka, H., Kobayashi, H., Kobayashi, A., Barzykin, V., and Gor'kov, L.P., *Phys. Rev. Lett*., **87**, 067002 (2001).
6. Jaccarino, V. and Peter, M., *Phys. Rev. Lett*., **9**, 290 (1962).
7. Larkin, A.I. and Ovchinnikov, Y.N., *Sov. Phys. JETP,* **20**, 762 (1962); Fulde, P. and Ferrell, R.A., *Phys. Rev*., **135**A, 550 (1964).